Chemical aging of atmospheric mineral dust during transatlantic transport

Reply to Anonymous Referee #3 (doi:10.5194/acp-2016-470-RC3)

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We thank the anonymous referee for the in-depth comments on this manuscript. The comments and questions raised are addressed below by our point-by-point reply (black) and the revised MS.

General comments

The abstract and the body of the text are not consistent, and the text does not efficiently support the conclusions in the abstract. In fact, the abstract and the text look like parts of different papers.
There are two major results in the abstract. One result is on the pattern of dust transport over the Atlantic, which is characterized by (1) a steep and linear westward gradient due to the dust sedimentation (dry deposition) in the DTA zone and (2) an efficient removal dominated by cloud interaction and wet deposition in the DIZ zone. Another result is on the aging process of dust particles and on the effect of the aging on dust AOD in addition to the removal of the dust. About the later result, authors give the details as (1) aging of dust particles by absorbing inorganic acids changes the particles into soluble modes, enhances the absorption of water vapor, and consequently causes the increase of AOD, which the authors name as "direct effect of dust aging", and (2) aging of dust particles causes more efficient removal of particles in comparison with non-aged dust particles, and consequently results in a decrease of dust AOD, which the authors name as "indirect effect of dust aging". However, the text of results and discussion in the manuscript does not focus on the above two results.

The abstract and the discussion in the manuscript have been revised accordingly.

Here are my understandings on the text. Section 3: In the first part (Figure 2, and also Figure 3, which is somewhat a repeat of Figure 2), the simulated result (first result mentioned above) and the possible reasons for the result are simply introduced and described. As a major result of this study described in the abstract, more details and a deep discussion are necessary. My major concern on this part is the lack of a discussion on the uncertainties in the result. Another concern is that this part is not consistent with the purpose of this Section, which is to evaluate the performance of the model (the first line of Section 3). The remaining parts of this section are the evaluation of the model performance with the comparison to AERONET observations.

The text has been revised to be consistent with the purpose of this section and a note on the

uncertainties of the result has been added. For a discussion on the uncertainties we refer, however, to Section 4 "Sensitivity studies", since this section is exactly about the modeling uncertainties.

Section 4: This part is an evaluation of model performance, too. First, the evaluation is conducted with a focus on the model sensitivity to emission flux and to removal mechanisms. Then the influence of different convection schemes and dust chemical aging on simulation results is examined. Although the major results described in the abstract are introduced in Section 3 and Section 4, the results are not described in a clear and compacted way. In addition, the explanations of the consistence and difference between the simulation results and the observational facts are very qualitative and the uncertainties are not quantitatively discussed.

The text has been revised such that this section 4 "Sensitivity studies", now clearly deals with modeling uncertainties (and not again of model performance evaluation).

The evaluation of the model performance is not bad and is acceptable. But the evaluation shows the quality of the model and has a weak relation with the conclusions described in the abstract.

The conclusions and the abstract have been revised accordingly.

So the contents of abstract are inconsistent with the contents of results and discussion (Section 3 and Section 4). Actually, many parts in the text of results and discussions are repeats of the paper of Abdelkader et al. (2015). The first result described in the abstract is original in this model study, but the second result contains less new information in comparison with Abdelkader et al. (2015).

The study of Abdelkader et al. (2015) presents the dust-air pollution interaction over the Easter Mediterranean, while this work focuses on a "Sensitivity of transatlantic dust transport to chemical aging and related atmospheric processes" – the new title (see our reply to referee #1). Since both studies focus on the chemical aging of dust, there is of course some overlap in the description. Otherwise this paper would not be able to stand alone. To our opinion, the overlap is small and important to have for the average reader to understand the main text flow without referring to Abdelkader et al. (2015), which an interested reader of course will/shall do.

Other major comments
The abstract is tedious and hardly followed.

The abstract has been revised.

Figure 1 is not necessary according to the abstract. The model has been described and evaluated in Abdelkader et al. (2015).

We prefer to have this paper a standalone (see our above) and, hence, we keep Figure 1.

Removal processes of dust particles by dry and wet deposition, including the subsequences of dust aging, are repeatedly applied to explain simulation results. In addition to that the repeats make the manuscript very tedious, almost all explanations lack of a discussion on the confidence of the explanations, i.e. to what a degree the explanations can account for the results. Discussions with quantitative evaluation are necessary to increase the quality of the explanations.

Redundancies have been removed and an extended discussion on a more quantitative evaluation has been included based on the statistical parameters shown in Table 1a,b of the Supplement.

The description on the wet deposition of dust particles associated with the aging of particles lacks of details and is not clear. The removal is simply described as the processes of the hygroscopic growth of aged particles (Section 4.3) and is discussed with comparisons associated with precipitation (convection) and dust emission (Section 4.2). Hygroscopic growth is a subsequence of particle aging (i.e. interaction with cloud), which is emphasized in this manuscript. However, precipitation is fundamentally governed by thermodynamic properties and the movement of air parcels (the convection: Aerosol particles are not included in the simulation of water vapor distributions by Tost et al. (2006b)). Precipitation removes dust particles via the adoption of dust particles by cloud droplets and raindrops in cloud and in below-cloud air (the effect of washout) and/or via the raincloud droplet formation on dust particles under saturate conditions in cloud or the adjacent air (the effect of nucleation scavenging). The two scavenging processes are closely dependent on the size of particles and droplets. Under saturate conditions (in cloud), dust-induced droplets (nucleation scavenging) may grow into a large droplets. But the size, rather than the composition, of a particle is the key factor for the nucleation at the size range of dust particles, usually larger than several hundred nano-meters (Dusek et al. 10.1126/science.1125261, Science, Vol. 312, Issue 5778, pp. 1375-1378). In below-cloud air under sub-saturate conditions, the growth of aged particles due to water vapor absorption is limited and the particles are not expected to frequently become considerable larger than the original particles. So the relative importance of the two processes in the dust removal needs to be clearly described and discussed in order to quantitatively show how important of the subsequence of dust aging is and how the aging enhances the removal of aged dust particles. It sounds that washout is not important for the removal of the dust particles in DIZ zone. Is this correct?.

No, the washout is of course also important for the removal of the dust particles in DIZ zone, but the chemical aging and scavenging of aged dust particles are according to our study more important in the DIZ–zone compared to DTA–zone. The text has been revised accordingly.

The definition of "direct effect" and "indirect effect" of dust aging needs to be carefully reconsidered. In this study, the effect is limited to that on AOD. However, there are many other effects associated with the aging, such as the absorption of acid gaseous species and the change of gas phase reactions. In addition, the definition may cause a confusion when readers think the "direct and indirect climate effects of aerosol particles".

We do agree that the "direct effect" and "indirect effect" of chemical aging of dust seems limited only by a definition of AOD, but it actually includes all other effects. Indeed, we try to limit the definition to the AOD, since only the net-effect AOD eventually drives the radiation. Of course, the total effect includes many other processes, such as heterogeneous reactions on dust particles, which can either increase of decrease the AOD. But, at the end of a computation step only the net-effect on AOD accounts. Therefore, we keep our definitions as introduced here.

References

Abdelkader, M., Metzger, S., Mamouri, R. E., Astitha, M., Barrie, L., Levin, Z., and Lelieveld, J.: Dust–air pollution dynamics over the eastern Mediterranean, Atmospheric Chemistry and Physics, 15, 9173–9189, doi:10.5194/acp-15-9173-2015, URL `http://www.atmos-chem-phys.net/15/9173/2015/`, 2015.